Thermal Alteration of Spores and Pollen and Maturity of Organic Matter of the Cretaceous System, Songliao Basin, Northeast China

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Abstract

The relationship between sporo-pollen color and the degree of maturation of organic matter is discussed with regard to oil generation and evolution, as typified by the Cretaceous system in the Daqing Oil Field, central Songliao Basin, Northeast China. Color variation of spores and pollen is considered as a function of sedimentary environment and thermal alteration. Sporo-pollen color is classified into seven grades, and the degree of thermal alteration is studied in terms of color index. Results show that the sporo-pollen color index for the strata at the depth of 1,000—3,000 m (stratigraphically from the first member of the Liangjiang Formation to the upper Quantou Formation) ranges from 2.5—5.0, corresponding to a palaeotemperature range of 60°—140°C. These are the optimum oil-generating strata. The strata underlying the lower Quantou Formation below 3,000 m with the color index in excess of 5 and the palaeotemperature over 140°C may be favorable for gas accumulation. As for the strata at the depth of less than 1,000 m, i.e., stratigraphically overlying the second member of the Liangjiang Formation, which are characterized by a color index of 1.0—1.5 and a palaeotemperature of less than 60°C, the degree of maturation of organic matter is lower than that in the oil-generating strata.

Introduction

Variations in color of spores and pollen grains in sedimentary rocks are closely related to thermal alteration. During post-diagenesis, spores and pollen grains in sediments must have undergone a series of changes in chemistry and structure with increasing burial depth, temperature and pressure, as is clarified by the color variation of spores and pollen from light to dark. Spores and pollen and other organic and inorganic materials in sediments seem to have the same fate in the process of thermal alteration, as a result of their simultaneous alteration under the same physico-chemical condition. Therefore, it is possible to predict the palaeotemperature and the degree of maturation of organic matter and to inquire into the mechanism of oil formation and evolution by studying thermal al teration of spores and pollen, coupled with the observation of their color variations, so as to provide the basis for the evaluation of oil-gas potential in a certain area.

In the Songliao Basin are widespread Gretaceous sedimentary strata, which are characterized by great thickness, complete stratigraphy, extremely abundant spores and pollen fossils and rich oils as evidenced by lots of drill core data available. Therefore, this basin is considered as the best candidiate for the study of the relationship between the color variation of spores and pollen and the degree of maturation of organic matter in sediments. This paper is intended to give a relatively detailed discussion of color changes of sporo-pollen fos-

sils in both vertical and horizontal directions,and their controlling factors on the basis of the available palynological data obtained from petroleum exploration in the Daqing Oil Field in the past two decades. Sporo-pollen color variation studies permit a deep understanding of the degree of maturation of organic matter, oil formation and evolution and the threshold depth and stratigraphic horizons favourble for oil-generation.

Color Index

Colors of spores and pollen grains in sedimentary rocks have been described by many workers. By observing visually the plant spores and cuticular plant debris from the strata in western Canada, F. L. Staplin[1] (1969) classified the color-change series from light to dark into five grades: fresh yellow; brownish yellow; brown black; and black, with additional evidence of rock metamorphism. Furthermore, he explicity related these grades to oil and gas occurrences: biogenic methane, oil, wet gas and condensate, dry gas, and dry gas to barren. The division of the color-change series of sporo-pollen fossils proposed by J. Cannan (1974)[2] involves six grades: yellow, light brown, brown, dark brown, very dark brown, and black. Statistical data on spores and pollen, in conjunction with observations of their colors, show that there are considerable amounts of pale yellow spores and pollen in sedimentary rocks, even at greater depths, in the Songliao Basin. Therefore, the above two schemes are not suitable for the present situation. Our scheme to describe the sporo-pollen colors includes seven grades: pale yellow, yellow, brownish yellow, brown, dark brown, brownish black, and black. However, the color of spores and pollen grains from the same sample often varies over a wide range. Generally, the influence factors are outlined as follows: (1) The thickness of exine, i.e., the thicker the exine, the darker the color. (2) The type of ornamentation. For example, the spores of *Lygodiaceae* are darker in color because of their thicker exines and well-developed ornamentation with spinulous, tumourous and ribbed forms. (3) Plant genus and species. For example, the pollen grains of pinaceae and *psilate triletes* are relatively light in color. (4) The concentration of organic matter in the sample. As spores and pollen grains are coated with carbonized organic matter, their color has been darkened. Although such organic matter can be removed by oxidizing agents, there is little doubt that it will make it more complicated to observe the colors of spores and pollen grains. (5) Sedimentary environment. A drill core sample, in fact, represents a small stratigraphical section because it has a certain thickness. The fact that dark and light laminae occur alternatively in some samples indicates differences in sedimentary environment and organic abundance. As a result, significant differences are frequently noticed in the color of spores and pollen. On account of many influence factors, the color of spores and pollen in the same sample may vary over a wide range. Thus, it is very difficult to describe definitely the colors of sporo-pollen fossils in a certain sample. The way to avoid these troubles is to make statistical observations on one or several species of sporo-pollen fossils. However, it is almost impossible to gain such a species of spor-pollen fossils as can be used for systematic correlation from Cretaceous strata, as thick as 6,000 m, in the Songliao Basin. Therefore, a statistical method of sporo-pollen color index has been adopted in this study. Generally, the following steps are taken: (a) to count the numbers of spores and pollen grains pertinent to each color grade in the sample, and (b) to calculate the weighted average of color grades of each sporo-pollen fossils, i.e., the sporo-pollen color index. For instance, the sporo-pollen color index of the

sample taken from Well Long-6 at the depth of 1,539.2 m (Table 1) has been calculated in the following way:

$$[(2 \times 6) + (3 \times 4) + (4 \times 2)] \div (6 + 14 + 2) = 2.8.$$

Before statistical observation of sporo-pollen colors, a set of sporo-pollen reference colors (from light to dark) should be prepared. In order to minimize artificial error this set of reference colors should always be used in practice to obtain as accurate statistical results as possible.

Table 1. Statistical data of sporo-pollen color index

Well No.	Depth (m)	Stratigraphy \ Number of sporo-pollen fossils \ Color grade	1 Pale yellow	2 Yellow	3 Brownish yellow	4 Brown	5 Dark brown	6 Very dark brown	7 Black	Color index	Principal color
Tong-1	266.0	First member of Mingshui Formation	4	11	4					2.0	Yellow
Long-6	1539.2	Second and third members of Qingshankou Formation		6	14	2				2.8	Brownish yellow
Sha-5	1758.58	Third member of Quantou Formation			2	8	18	5		4.8	Dark brown

Sporo-pollen Color and Sedimentary Environment

Spores and pollen grains preserved under different sedimentary conditions are different in color. This indicates that the differences in color for various sporo-pollen fossils are closely related to sedimentary environments. With respect to the preservation and color variation of spores and pollen grains in Cretaceous strata of the Songliao Basin, two principal mechanisms——"petrification" and " carbonization" are proposed.

The so-called "petrification" refers to the changes spores and pollen have undergone under oxidizing conditions after they were preserved in sediments. Organic matter, for the most part, is destroyed in the process of oxidation and therefore, only small amounts of spores and pollen have been preserved as fossils in sediments and cemented by calcareous and siliceous materials, although the exines of spores and pollen grains are very firm. The host rocks, usually red or green, contain a few poorly-preserved sporo-pollen fossils. With increasing depth of burial, geothermal temperatures ries and thermal alteration becomes intense and thus the color of spores and pollen varies from light yellow to greenish grey. Finally, the fossils may be destroyed completely. Statistical data show that the petrified spores and pollen grains are common in the strata above the depth of 2,500 m in the Songliao Basin, corresponding to the strata overlying the secon memeber of the Qauntou Formation, whereas spores and pollen grains are rare below 2,500 m.

"Carbonization" refers to the changes spores and pollen grains have undergone under reducing conditions. A significant amount of organic matter, together with spores and pollen,

is preserved under reducing conditions. With increasing depth of burial, temperature and intensity of thermal alteration, the exines of spores and pollen grains are gradually carbonized, leading to the color change from light to dark, i.e., from pale yellow to black. Sporo-pollen fossils surrounded by more severely "carbonized" organic matter are still darker in color. Sediments deposited under reducing conditions usually contain abundant, well-preserved, highly transparent spores and pollen grains. With intensifying thermal alteration, the transparency of sporo-pollen fossils tends to decrease and their color becomes darker and darker gradually. Eventually, they would be converted into totally carbonized particles, even into graphite. In the central part of the Songliao Basin, the Cretaceous strata below 3,800 m (corresponding to the strata underlying the Denglouku Formation) contain 25—30% black spores and pollen grains with the rest being dark brown to brownish black.

Table 2. Comparison between petrified and carbonized spores and pollen

Item	Petrification	Carbonization
Geochemical environment	Oxidation	Reduction
Oxidizing degradation	Strong	Weak or no
Thermal degradation	Less remarkable	Strong
Preservation of sporo-pollen fossils	Poor, in minor amounts	Good, in large amounts
Sporo-pollen transparency	Poor	Good (→poor)
Sporo-pollen color	Pale yellow, greenish grey	Pale yellow to black
Host rock color	Red, green	Grey, very dark grey, black
Chemical composition of cements	Calcareous, siliceous	Carbonaceous, calcareous or siliceous
Sedimentary facies zone	Delta branch-plain, littoral, etc.	Swamp, delta front, shallow-water, semi-deep-and deep-water
Oil-gas generation	Poor or barren	Oil-precursor

Since the preservation of sporo-pollen fossils and their color changes are controlled by geochemical conditions, spores and pollen grains from sediments deposited in different sedimentary environments must have their own distribution characteristics. The planar distribution pattern of sporopollen fossils from the first member of the Yaojia Formation is shown in Fig. 1. From Fig. 1 the preliminary conclusions can be drawn as follows:

(1) The content of spores and pollen grains in sedimentary rocks increases gradually from the margin to the centre in the Songliao Basin. (2) The lake deposits contain abundant spores and pollen grains, whereas the onshore deposits contain fewer or even no spores and pollen. (3) As viewed from their distribution in sedimentaryfacies-zones, spores and pollen are absent in the sediments of alluvial, flood-plain and piedmont-alluvial-plain facies, rare in the sediments of delta branch-plain and littoral facies (accounting for 10—20% of the sample analyzed). most abundant in the sediments of delta front and shallow-water facies, and relatively abundant in the sediments of semi-deep and deep water facies (amounting to about 80% of the sample analyzed). (4) The sediments of delta branch-plain and littoral facies generally contain minor spores and pollen grains, and the sporo-pollen assemblages are relatively simple. In the sediments of delta front facies, sporo-pollen fossils are very abundant, and the sporo-pollen assemblages are complex. Spores and pollen grains in the sediments of deep water facies are relatively abundant, and the sporo-pollen assemblages are rather simple. (5) Spores and pollen grains from the sediments of delta branch-plain and lit-

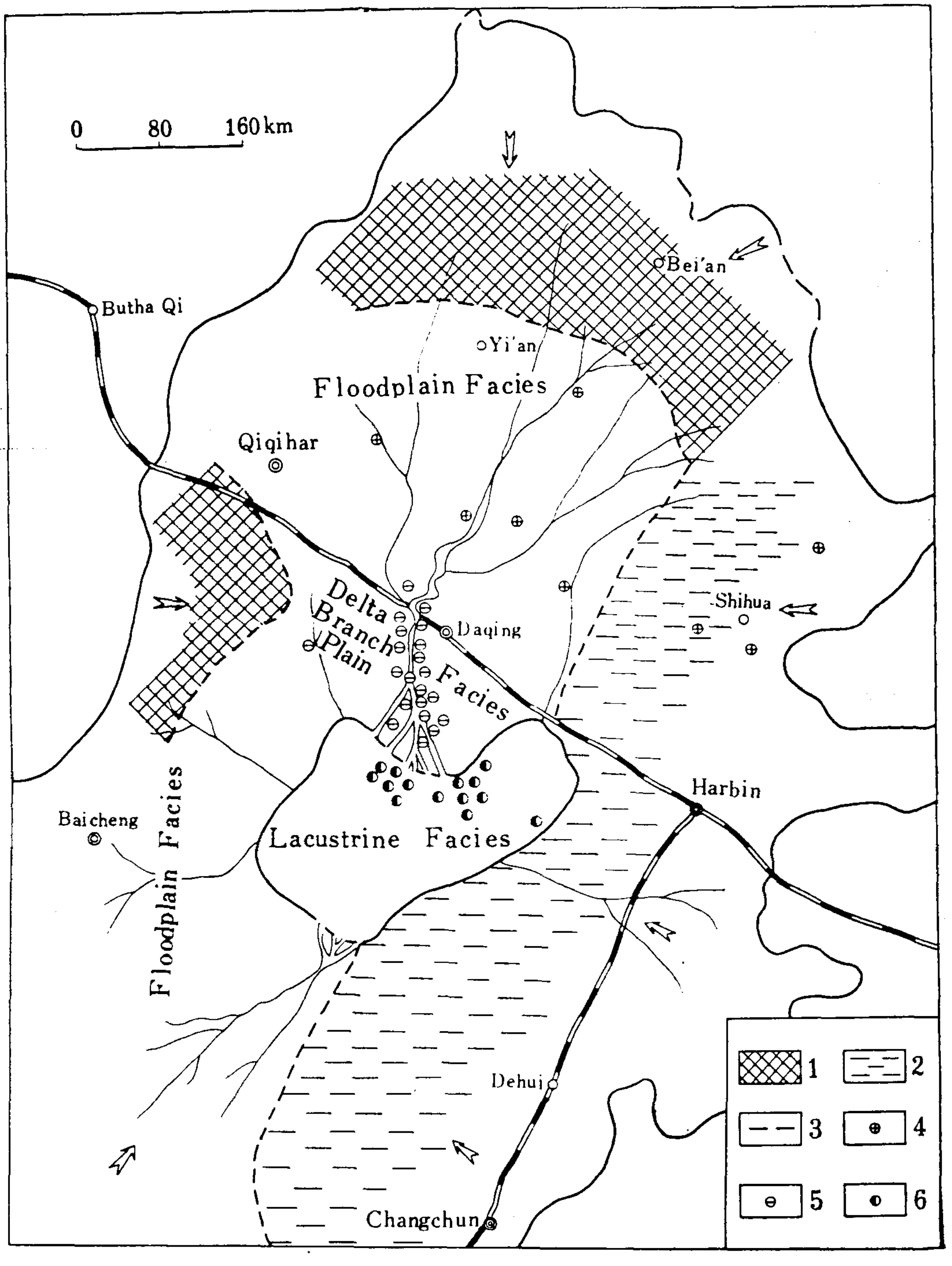

Fig. 1. Sporo-pollen distribution in different sedimentary facies of the Yiaojia Formation in the Songliao Basin.

1. Eroded area; 2. piedmont plain facies; 3. facies boundary; 4. well site without sporo-pollen fossils; 5. well site with minor sporo-pollen fossils; 6. well site with abundant sporo-pollen fossils.

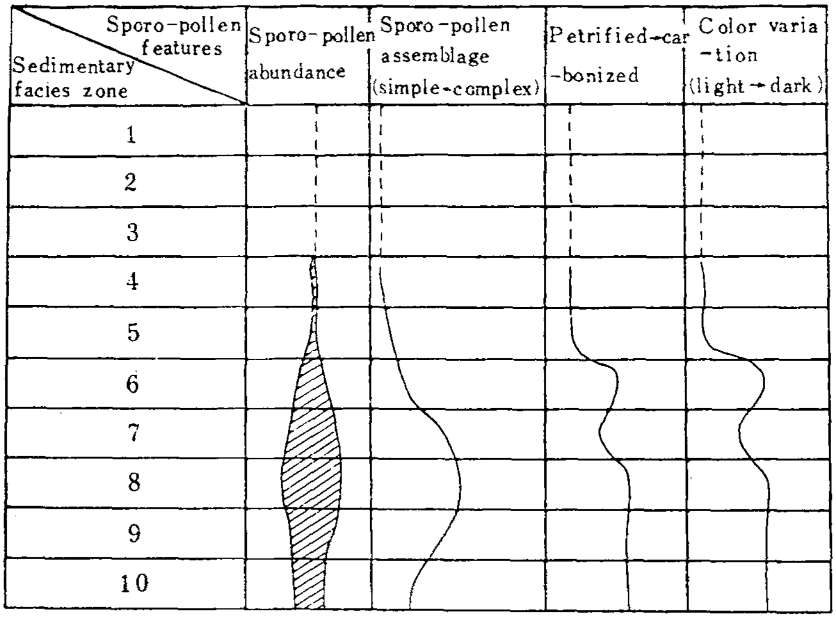

Fig. 2. The relationship between sporo-pollen color and sedimentary facies in the Songliao Basin.

1. Alluvial; 2. floodplain; 3. piedmont plain; 4. delta branch-plain; 5. beach littoral; 6. swamp; 7. shallow-water; 8. delta front; 9. semi-deep-water; 10. deep-water.

toral (except swamp) facies usually have been subjected to petrification and thus their color is lighter, while those preserved in the sediments of delta front, semi-deep——deep water and swamp facies have undergone carbonization and, therefore, their color is darker. Owing to varying sedimentary conditions in the shallow-water environment——a weak oxidation-reduction geochemical environment, spores and pollen grains are of diverse source. They are transported by streams and broocklets and deposited in the shallow water environment, where the majority of them have been carbonized and partly petrified. Therefore, the color of spores and pollen grains varies over a wide range.

The sediments of delta branch-plain sub-facies may have undergone swamping, and those of littoral sub-facies may contain abundant, darker spores and pollen grains due to carbonization. For example, samples taken from the two sub-facies mentioned above in the first member of the Yiaojia Formation contain 1/6—1/7 sporo-pollen fossils. In summary, the preservation of spores and pollen and their subsequent color changes depend on both sedimentary environment and burial depth (palaeogeothermal temperature). In orther words, sedimentary environment is a decisive control on whether spores and pollen can be preserved and whether they will either be petrified or carbonized following the preservation, while burial depth has a great bearing on the severity of thermal alteration and the degree of color variation of spores and pollen.

The petrified spores and pollen occur in small amounts and no obvious changes in color are noticed with intensifying thermal alteration. For this reason, carbonized spores and pollen are of key importance in the study of thermal alteration of spores and pollen and in the establishment of the relationship of sporo-pollen color to the degree of maturation of organic matter.

Sporo-pollen Color Index and Palaeogeothermal Temperature

Statistical data on sporo-pollen color index for more than 1,000 drill core samples from 23 wells in the Songliao Basin are presented in Table 3. Perusal of this table shows a general trend of increasing color index with burial depth. It is confirmed that the majority of spores and pollen in the same sample show no difference in color, although in some cases the color of some spores and pollen grains varies greatly. What is more peculiar is that spores and pollen at greater depths are lighter in color than those at shallower depths in the same stratigraphical unit.

Table 3. Variations in color of sporo-pollen fossils at various depths of the Songliao Basin

Depth (m)	Sporo-pollen color index	Sporo-pollen color	
		Principal color	Range
<100	1.1	Pale yellow	Pale yellow to yellow
100—500	1.8	Yellow	Pale yellow to brownish yellow
500—1,000	2.5	Yellow to brownish yellow	Pale yellow to brown
1,000—1,500	3.0	Brownish yellow	Yellow to dark brown
1,500—2,000	4.5	Brown to dark brown	Yellow to very dark brown
2,000—3,000	5.1	Dark brown	Brownish yellow to black
3,000—4,000	5.8	Very dark brown	Brown to black
>4,000	>6.2	Very dark brown	Dark brown to black

Table 4. Sporo-pollen color and degree of maturation of organic matter of Cretaceous strata in the Songliao Basin

Formation	Depth (m)	Color index	Sporo-pollen color	Vitrinite reflectance (%)	Palaeogeo-thermal condition	Maturity of organic matter (evolutionary stage)	Oil-gas occurrence
Mingshui	<100 100—500	1.1—1.8	Pale yellow to yellow	0.3	<60℃	Non-maturation	Biogenic methane
Sifangtai	500—1,000	2.5	Yellow to brownish yellow	0.3—0.5			
Liangjiang	1,000—1,500	2.5—4.5	Brownish yellow to dark brown	0.5—0.9	60—110℃	Lower maturation	Heavy oil
Yaojia	1,500—2,000						
Qingshankou	2,000—3,000	5.0	Dark brown	0.9—1.3	110°—140℃	Higher maturation	Light oil
Quantou	3,000—4,000	5.0—5.8	Dark brown to very dark brown	1.3—2.0	140°—170℃		Wet gas
Denglouku	>4,000	6.2—7.0	Very dark brown to black	>2.0	>170℃	Super-maturation	Dry gas

This anomaly may be reasonably explained by the occurrence of petrified spores and pollen. Therefore, the author suggests that pale yellow spores and pollen occurring in the strata below 1,000 m in the Songliao Basin should not be taken into consideration in statistical treatment. All sporo-pollen fossils above 1,000 m are relatively light in color and consequently it is difficult to distinguish petrified spores and pollen from carbonized ones. On this condition it is recommended to take all spores and pollen into account.

The color change of spores and pollen in sediments depends mainly on palaeogeothermal conditions. Generally, the higher the palaeogeothermal temperature, the darker the color of spores and pollen in sediments. The reason may be that the exines of spores and pollen are getting darkened due to strong carbonization in response to intensifying thermal alteration with increasing temperatures. It is commonly accepted that palaeogeothermal conditions may be affected not only by burial depth, but also by many other factors, such as lithological character of the basement, fault, depth of the Moho, etc. With the development of geological history and continuation of tectonic movements, the present burial depths and temperatures may be quite different from the past noes for a certain formation. However, sporo-pollen colors may truly reflect the maximum palaeogeothermal temperature or the maximum burial depth. J. Brooks and G. Shaw[3] considered the sporo-pollen color change as an indicator of the maximum palaeogeothermal temperature to which sediments were subjected. So sporo-pollen color studies can provide insight into the fluctuation of palaeogeothermal temperatures.

In predicting palaeogeothermal temperatures, the sporo-pollen color index must be used only in conjunction with other parameters such as vitrinite reflectance (VR), electron spin resonance (ESR), etc.

On the basis of the sporo-pollen color index, coupled with the data on vitrinite reflectance, the author has established the relationship of color index of Cretaceous spores and pollen to palaeogeothermal temperature of the Songliao Basin. When the sporo-pollen color index ranges from 1 to 2.5 or the Ro value (VR) from 0.3% to 0.5%, the corresponding palaeogeothermal temperature is less than 60°C; when the color index varies from 2.5—4.5 or the Ro value is in the range 0.9—0.9%, the corresponding palaeogeothermal temperature ranges from 60°—110°C; when the index is about 5 or Ro is in the range 0.9—1.3%, the corresponding palaeogeothermal temperature lies between 110°—140°C; and when the index > 5 or $Vo > 1.3$, the corresponding palaeogeothermal temperature is over 140°C (Table 4).

Sporo-pollen Color Index and Degree of Maturation of Organic Matter

In general, organic materials in sedimentary rocks are composed of soluble organic matter and kerogen. Kerogen consists chiefly of "inert matter", vitrinite, sporinite and sapropelic material. At a moderate degree of maturation, sapropelic material and sporinite can be converted into liquid hydrocarbons, while vitrinite can produce only dry gas. Spores and pollen are the important components of organic materials in sedimentary rocks, and also the highly productive oil-precursors. Since the color change and the severity of thermal alteration of

spores and pollen can directly mirror the degree of maturation of organic matter and also have close relations with oil generation and evolution, it is possible to make a proper evaluation of source rocks and oil-gas prospects in a certain district on the basis of the degree of maturation of organic matter, in combination with other important geochemical data.

On the basis of the sporo-pollen color index, in conjunction with data on vitrinite reflectance, the author has estimated palaeogeothermal temperature and degree of maturation of organic metter, and also evaluated the prospect of Cretaceous oil and gas in the Songliao Basin (Table 4). The studies of physico-chemical changes of some minerals in rocks provide further evidence for the classification of maturation stages of organic matter and of oil-gas evolution stages, as is shown in Table 4.

The data of Table 4 indicate that the sporo-pollen color index at the depth of 1,000—3,000 m (stratigraphically from the first member of the Liangjiang Formation to the upper Quantou Formation) ranges from 2.5 to 5, corresponding to a palaeogeothermal temperature of 60°—140°C. At this depth interval the maturity of organic matter is at moderate to high levels. So the strata there are the most important oil prospect targets. Below 3,000 m, the strata underlying the lower Quantou Formation with the color index in excess of 5 and the palaeogeothermal temperature over 140°C are prospects for gas, because organic materials there have undergone strong thermal alteration, so that they have become highly mature or even supermature. The strata above 1,000 m, stratigraphically overlying the second member of the Liangjiang Formation, have a color in dex of 1—2.5, corresponding to the palaeogeothermal temperature below 60°C, indicating that organic matter is not mature enough to convert into petroleum. Therefore, during this evolutionary stage, organic matter yields predominantly methane through biochemical processes.

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